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Generalized Expressions of Effective Nonlinear Optical Coefficient for Non-collinear Phase Matching in Uniaxial and Cubic Media

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Abstract

The conditions for non-collinear phase matched frequency conversion are analyzed and the corresponding expressions of the effective nonlinear optical coefficient(d_{eff}) for 13 classes of uniaxial crystals and 3 classes of cubic crystals are derived. The discussed cases correspond to the situation when $d_{ijk} \neq d_{ikj}$, when $d_{ijk} = d_{ikj}$, and when Kleinman symmetry condition holds, with the general consideration that the extraordinary ray(e-ray) is not perpendicular to the phase propagation vector **k** in the uniaxial medium.

Key words: d_{eff} , non-collinear phase matching.

I. Introduction

Non-collinear phase matching can be a very useful frequency conversion method in experiments of nonlinear optics[1,2,3,4]. It is used in the measurement of the duration of ultrafast pulses with background free autocorrelation between pulses[5,6]. It also has some advantage over collinear phase matching by providing more flexibility. Unlike collinear phase matching, the configuration of the propagation directions of each light wave in non-collinear phase matching is not unique. It can provide more degrees of freedom for optimizing the frequency conversion. With different configurations of the light wave propagation directions, non-collinear phase matching could also be used to characterize each $\chi^{(2)}$ component of a crystal. In particular, non collinear phase matching could occur in cubic media, where collinear phase matching is impossible due to the isotropy of refractive indices. More interestingly, phase matched frequency conversion from e-ray + e-ray $\rightarrow$ e-ray and o-ray + o-ray $\rightarrow$ o-ray can be achieved through non-collinear phase matching(o-ray means ordinary ray). These processes are disallowed in collinear phase matching.

The d_{eff} expressions for collinear phase matching in 13 uniaxial crystal classes were originally given by Zernike and Midwinter[1]. They have been modified by us in an earlier paper with the general consideration that e-ray is not perpendicular to the phase propagation vector **k** and the last two indices of d_{ijk} are not commutable[7]. It is important to derive the d_{eff} expressions for non-collinear phase matching with the same considerations, as they have relevance to experiments.

In part II, the definition of d_{eff} is reviewed and the conditions of non-collinear phase matching are discussed. In part III, the d_{eff} expressions for the

cases of $d_{ijk} \neq d_{ikj}$ and $d_{ijk} = d_{ikj}$, and for the media with Kleinman symmetry, are given separately. These expressions can also be used for non phase matched frequency conversion, provided that the phase matching equations in part II are not used.

II. Definition of d_{eff} and the Requirements for Non-collinear Phase Matching

Let us consider two waves $\mathbf{E}_1 = \mathbf{e}_1 A_1 \exp(i\mathbf{k}_1 \cdot \mathbf{r} - \omega_1 t)$ and $\mathbf{E}_2 = \mathbf{e}_2 A_2 \exp(i\mathbf{k}_2 \cdot \mathbf{r} - \omega_2 t)$ interacting inside the nonlinear crystal and generating the third wave $\mathbf{E}_3 = \mathbf{e}_3 A_3 \exp(i\mathbf{k}_3 \cdot \mathbf{r} - \omega_3 t)$. The frequency conversion is described by the coupled wave equation[7,8].

$$\frac{dA_3}{dz'} = 4 \frac{CA_1A_2}{\cos\beta + \sin\xi_3 \cos\alpha_3} d_{eff} \exp(i\Delta\mathbf{k} \cdot \mathbf{r}) \qquad \text{(II.1)}$$

for $\omega_1 \neq \omega_2$ or $\mathbf{e}_1 \neq \mathbf{e}_2$ or $\mathbf{k}_1 \neq \mathbf{k}_2$.

$$\frac{dA_3}{dz'} = 2 \frac{CA_1A_2}{\cos\beta + \sin\xi_3 \cos\alpha_3} d_{eff} \exp(i\Delta\mathbf{k} \cdot \mathbf{r}) \qquad \text{(II.2)}$$

for $\omega_1 = \omega_2$ and $\mathbf{e}_1 = \mathbf{e}_2$ and $\mathbf{k}_1 = \mathbf{k}_2$;

where $\Delta\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3$ and $C = i\dfrac{4\pi\omega_3^2}{2k_3c^2}$. z' is the amplitude attenuation direction, normal to the constant amplitude plane (the constant amplitude plane is usually parallel to the surface of the nonlinear medium). ξ_3 is the angle between S_3(Poynting vector) and $\mathbf{k}_3$ of the generated frequency ω_3. If ω_3 is an o-ray, then $\xi_3 = 0$. α_3 is the angle between z' and $\mathbf{e}_3$ (polarization of ω_3). β is the angle

between z' and $\mathbf{k}_3$. These parameters are shown in Figure 1. The optical frequencies should satisfy $\omega_1 + \omega_2 = \omega_3$. When $\omega_1 > 0$ and $\omega_2 > 0$ and $\omega_3 > 0$, we have sum frequency generation. The case of $\omega_1 > 0$ and $\omega_2 < 0$ and $\omega_3 > 0$, on the other hand, denotes difference frequency generation. $\mathbf{k}_i = \frac{n_i \omega_i}{c} \hat{k}_i$, where $\hat{k}_i$ is the unit vector of the phase propagation direction for each wave. d_{eff} is defined by the following equation;

$$d_{eff} = \sum_{ijk} e_{3i} d_{ijk} e_{1j} e_{2k}$$

$$= \mathbf{e}_3 \cdot [(\mathbf{d}^{[3]} \cdot \mathbf{e}_2) \cdot \mathbf{e}_1] \qquad \text{(II.3)}$$

where $\mathbf{d}^{[3]}$ denotes a third rank tensor.

When phase matching condition is satisfied, i.e. $\Delta\mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3 = 0$, the frequency conversion will be maximized. To satisfy this relation, $k_1 = \frac{n_1 \omega_1}{c}$, $|k_2| = \frac{n_2 |\omega_2|}{c}$ and $k_3 = \frac{n_3 \omega_3}{c}$ should be able to form 3 sides of a triangle and the following equation should be satisfied.

$$\frac{|n_1 \omega_1 - n_2 |\omega_2||}{\omega_1 + \omega_2} < n_3 < \frac{n_1 \omega_1 + n_2 |\omega_2|}{\omega_1 + \omega_2} \qquad \text{(II.4)}$$

This requirement is sufficient for non-collinear phase matching in cubic media(isotropic refractive index) and for the case of o-ray + o-ray →o-ray, since the phase matching condition can be achieved by adjusting the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$ while keeping each refractive index unchanged. For example, one can perform phase matched non-collinear second harmonic generation in a cubic medium as long as $n(\omega) > n(2\omega)$ is satisfied. However, if any e-ray is involved, this requirement is not adequate since the refractive index of the e-ray is dependent on the wave propagation direction.

Let us define each wave propagation direction with angles θ, ϕ as shown in Figure 2, and suppose θ_1 and θ_2 of the fundamental frequencies and ϕ_3 of the generated frequency are known. We want to find out the conditions for phase matched frequency conversion and the corresponding values of ϕ_1 and ϕ_2 and θ_3. We can use $\Delta\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3=0$ to obtain the following equations:

$$n_1\omega_1\cos\theta_1+n_2\omega_2\cos\theta_2=n_3\omega_3\cos\theta_3 \tag{II.5}$$

$$n_1\omega_1\sin\theta_1\cos\phi_1+n_2\omega_2\sin\theta_2\cos\phi_2=n_3\omega_3\sin\theta_3\cos\phi_3 \tag{II.6}$$

$$n_1\omega_1\sin\theta_1\sin\phi_1+n_2\omega_2\sin\theta_2\sin\phi_2=n_3\omega_3\sin\theta_3\sin\phi_3 \tag{II.7}$$

If ω_3 is an e-ray, n_3 is described by:

$$\frac{1}{n_3^2}=\frac{\cos^2\theta_3}{n_{3o}^2}+\frac{\sin^2\theta_3}{n_{3e}^2} \tag{II.8}$$

where n_{3o} and n_{3e} are the principle values of the ordinary and extraordinary refractive index at ω_3.

The solutions are:

$$n_3=n_{3e}\sqrt{1-\frac{n_{eff}^2}{n_{3o}^2}+\frac{n_{eff}^2}{n_{3e}^2}} \tag{II.9}$$

where $n_{eff}=n_1\dfrac{\omega_1}{\omega_3}\cos\theta_1+n_2\dfrac{\omega_2}{\omega_3}\cos\theta_2$ (II.10)

$$\cos\theta_3=\frac{n_{eff}}{n_{3e}\sqrt{1-\dfrac{n_{eff}^2}{n_{3o}^2}+\dfrac{n_{eff}^2}{n_{3e}^2}}} \tag{II.11}$$

$$\cos(\phi_1-\phi_3)=\frac{A_3^2+A_1^2-A_2^2}{2A_3A_1} \tag{II.12}$$

$$\cos(\phi_2 - \phi_3) = \frac{A_3^2 + A_2^2 - A_1^2}{2A_3A_2} \qquad \text{(II.13)}$$

where $A_i = n_i\omega_i \sin\theta_i$. We can choose $0 \le \phi_1 - \phi_3 \le \pi$ for (II.12), $-\pi \le \phi_2 - \phi_3 \le 0$ for (II.13) when $\omega_2>0$, and $0 \le \phi_2 - \phi_3 \le \pi$ for (II.13) when $\omega_2<0$.

To ensure that $|\cos\theta_3| < 1$, we need:

$$|n_{eff}| < n_{3o} \qquad \text{(II.14)}$$

To give a reasonable result for ϕ_1 and ϕ_2, we need $|A_1|$, $|A_2|$ and $|A_3|$ form 3 sides of a triangle. This is guaranteed by (II.4), since $\frac{|A_1|}{c}$, $\frac{|A_2|}{c}$, $\frac{|A_3|}{c}$ are the projections of $\mathbf{k}_1$, $\mathbf{k}_2$ and $\mathbf{k}_3$ on the xy plane.

If ω_3 is an o-ray, we need to use;

$$n_3 = n_{3o} \qquad \text{(II.15)}$$

and $$\cos\theta_3 = \frac{n_{eff}}{n_{3o}} \qquad \text{(II.16)}$$

to substitute (II.9) and (II.11). Other relations (II.10), (II.12), (II.13), and (II.14) remain the same.

(II.4) and (II.14) are the requirements for non-collinear phase matching in uniaxial media when any e-ray is involved.

III. d_{eff} Expressions

Unlike collinear phase matching, non-collinear phase matching can be

achieved through e-ray + e-ray →e-ray and o-ray + o-ray →o-ray, since there is more flexibility for adjustment. Therefore, we derive the d_{eff} expressions of the following six cases for both uniaxial and cubic media.

Case (1): $\mathrm{o-ray}(\omega_1) + \mathrm{o-ray}(\omega_2) \rightarrow \mathrm{e-ray}(\omega_3)$

$\mathbf{e}_1 = (\sin\phi_1, -\cos\phi_1, 0)$

$\mathbf{e}_2 = (\sin\phi_2, -\cos\phi_2, 0)$

$\mathbf{e}_3 = (-\cos\theta_{s3} \cos\phi_3, -\cos\theta_{s3} \sin\phi_3, \sin\theta_{s3})$

where ϕ_i is the azimuthal angle for the ith wave propagation direction, θ_{si} is the angle between the optical axis and the Poynting vector of the ith wave, which relates to the wave propagation angle θ_i by: $\tan\theta_{si} = \frac{n_{io}^2}{n_{ie}^2} \tan\theta_i$.

Case (2): $\mathrm{e-ray}(\omega_1) + \mathrm{e-ray}(\omega_2) \rightarrow \mathrm{o-ray}(\omega_3)$

$\mathbf{e}_1 = (-\cos\theta_{s1} \cos\phi_1, -\cos\theta_{s1} \sin\phi_1, \sin\theta_{s1})$

$\mathbf{e}_2 = (-\cos\theta_{s2} \cos\phi_2, -\cos\theta_{s2} \sin\phi_2, \sin\theta_{s2})$

$\mathbf{e}_3 = (\sin\phi_3, -\cos\phi_3, 0)$

Case (3): $\mathrm{o-ray}(\omega_1) + \mathrm{e-ray}(\omega_2) \rightarrow \mathrm{e-ray}(\omega_3)$

$\mathbf{e}_1 = (\sin\phi_1, -\cos\phi_1, 0)$

$\mathbf{e}_2 = (-\cos\theta_{s2} \cos\phi_2, -\cos\theta_{s2} \sin\phi_2, \sin\theta_{s2})$

$\mathbf{e}_3 = (-\cos\theta_{s3} \cos\phi_3, -\cos\theta_{s3} \sin\phi_3, \sin\theta_{s3})$

Case (4): $\mathrm{o-ray}(\omega_1) + \mathrm{e-ray}(\omega_2) \rightarrow \mathrm{o-ray}(\omega_3)$

$\mathbf{e}_1 = (\sin\phi_1,\ -\cos\phi_1,\ 0)$

$\mathbf{e}_2 = (\ -\cos\theta_{s2}\cos\phi_2,\ \ -\cos\theta_{s2}\sin\phi_2,\ \ \sin\theta_{s2}\ )$

$\mathbf{e}_3 = (\sin\phi_3,\ -\cos\phi_3,\ 0)$

Case (5): $\mathrm{e-ray}(\omega_1) + \mathrm{e-ray}(\omega_2) \rightarrow \mathrm{e-ray}(\omega_3)$

$\mathbf{e}_1 = (\ -\cos\theta_{s1}\cos\phi_1,\ \ -\cos\theta_{s1}\sin\phi_1,\ \ \sin\theta_{s1}\ )$

$\mathbf{e}_2 = (\ -\cos\theta_{s2}\cos\phi_2,\ \ -\cos\theta_{s2}\sin\phi_2,\ \ \sin\theta_{s2}\ )$

$\mathbf{e}_3 = (\ -\cos\theta_{s3}\cos\phi_3,\ \ -\cos\theta_{s3}\sin\phi_3,\ \ \sin\theta_{s3}\ )$

Case (6): $\mathrm{o-ray}(\omega_1) + \mathrm{o-ray}(\omega_2) \rightarrow \mathrm{o-ray}(\omega_3)$

$\mathbf{e}_1 = (\sin\phi_1,\ -\cos\phi_1,\ 0)$

$\mathbf{e}_2 = (\sin\phi_2,\ -\cos\phi_2,\ 0)$

$\mathbf{e}_3 = (\sin\phi_3,\ -\cos\phi_3,\ 0)$

For a cubic crystal, there is no difference between e-ray and o-ray, because its three principle values of refractive index are equal. If the phase matching condition is satisfied for one of the above cases, it should also be satisfied for the rest 5 cases. Therefore, the interference among the outcoming e-rays and o-rays from all 6 cases should be considered.

The last two indices of d_{ijk} are generally not commutable. Therefore, the d_{eff} definition (II.3) is used to derive the d_{eff} expressions for each crystal. The nonvanishing elements of each crystal are from reference [9], [10] and [11].

By comparing the d_{eff} definitions for case(2) and case(3),

$$d_{eff}(\text{case}(2)) = \sum_{ijk} d_{ijk} e_i(\omega_3\ o-\text{ray})\, e_j(\omega_1\ e-\text{ray})\, e_j(\omega_2\ e-\text{ray}) \tag{III.1}$$

$$d_{eff}(\text{case}(3)) = \sum_{ijk} d_{ijk} e_i(\omega_3\ e-\text{ray})\, e_j(\omega_1\ o-\text{ray})\, e_j(\omega_2\ e-\text{ray})$$

$$= \sum_{ijk} d_{jik} e_i(\omega_1\ o-\text{ray})\, e_j(\omega_3\ e-\text{ray})\, e_j(\omega_2\ e-\text{ray}) \tag{III.2}$$

we can obtain the d_{eff} expressions for case(3) from the d_{eff} expressions for case(2) by exchanging the first two indices of d_{ijk} and changing ω_1, θ_1, ϕ_1 to ω_3, θ_3, ϕ_3 and ω_3, θ_3, ϕ_3 to ω_1, θ_1, ϕ_1. Similarly, we can obtain the d_{eff} expressions for case(4) from the d_{eff} expressions for case(1) by exchanging the first one and last one indices of d_{ijk} and changing ω_2, θ_2, ϕ_2 to ω_3, θ_3, ϕ_3 and ω_3, θ_3, ϕ_3 to ω_2, θ_2, ϕ_2.

The d_{eff} expressions for cases of $d_{ijk} \neq d_{ikj}$ are given in Tables 1(a), 2(a), 3(a), 4(a), 5(a), and 6.

When sum frequency generation is performed and the two fundamental frequencies are close to each other, the difference between d_{ijk} and d_{ikj} is very small and can be neglected. For second harmonic generation, the relation $d_{ijk} = d_{ikj}$ is strictly valid. Therefore, we can obtain the d_{eff} expressions for cases of $d_{ijk} = d_{ikj}$ from the above results for $d_{ijk} \neq d_{ikj}$. In these cases, the last two indices j and k in d_{ijk} can be contracted to ℓ ($d_{i\ell} = d_{ijk}$) according to the following rule:

$$xx \to 1,\ yy \to 2,\ zz \to 3,\ yz \text{ or } zy \to 4,\ zx \text{ or } xz \to 5,\ xy \text{ or } yx \to 6 \tag{III.3}$$

The d_{eff} expressions for cases of $d_{ijk} = d_{ikj}$ and for cases when Kleinman symmetry holds are given in Tables 1(b), 2(b), 3(b), 4(b), 5(b), 5(c), and 6.

For non phase matched frequency conversion, all the results in Table 1 through Table 6 are usable provided that the phase matching equations in part II

are not used.

IV. Summary

The conditions for non-collinear phase matched frequency conversion are analyzed and the corresponding d_{eff} expressions for 13 classes of uniaxial crystals and 3 classes of cubic crystals are derived. The discussed cases correspond to the situation when $d_{ijk} \neq d_{ikj}$, when $d_{ijk} = d_{ikj}$, and when Kleinman symmetry condition holds, with the general consideration that e-ray is not perpendicular to the phase propagation vector **k** in an uniaxial medium. These equations and d_{eff} expressions can be used to optimize frequency conversion in practical applications and characterize each component of d_{ijk} in the experiment.

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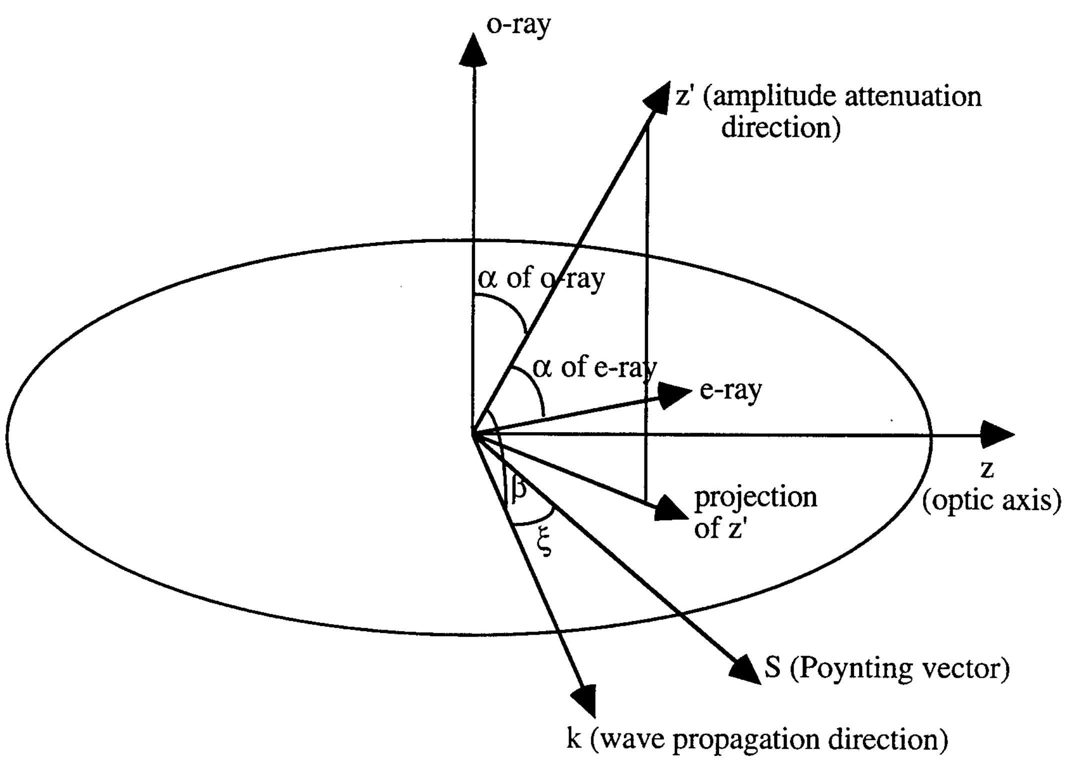

Figure 1. Physical meaning of the parameters in the coupled wave equation.

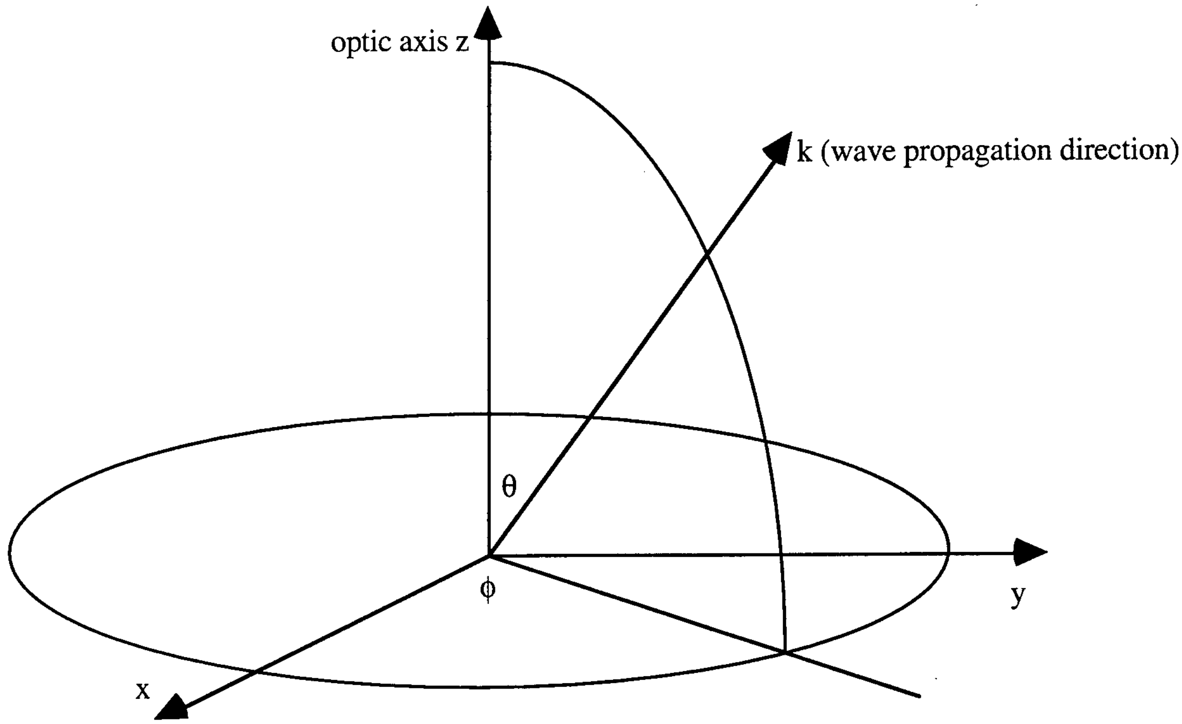

Figure 2. Wave propagation direction in a crystal.

Table 1(a). d_{eff} expressions for case (1): $o-ray(\omega_1)+o-ray(\omega_2)\rightarrow e-ray(\omega_3)$, in the case of $d_{ijk}\neq d_{ikj}$, and without the Kleinman symmetry condition.

Crystal class	d_{eff} without the Kleinman symmetry condition
6 and 4	$(d_{zxx}\cos(\phi_1-\phi_2)-d_{zxy}\sin(\phi_1-\phi_2))\sin\theta_{s3}$
622 & 422	$-d_{zxy}\sin\theta_{s3}\sin(\phi_1-\phi_2)$
6mm & 4mm	$d_{zxx}\sin\theta_{s3}\cos(\phi_1-\phi_2)$
$\bar{6}m2$	$-d_{yyy}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$
3m	$d_{zxx}\sin\theta_{s3}\cos(\phi_1-\phi_2)-d_{yyy}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$
$\bar{6}$	$(d_{xxx}\cos(\phi_1+\phi_2+\phi_3)-d_{yyy}\sin(\phi_1+\phi_2+\phi_3))\cos\theta_{s3}$
3	$[d_{xxx}\cos(\phi_1+\phi_2+\phi_3)-d_{yyy}\sin(\phi_1+\phi_2+\phi_3)]\cos\theta_{s3}$ $+[d_{zxx}\cos(\phi_1-\phi_2)-d_{zxy}\sin(\phi_1-\phi_2)]\sin\theta_{s3}$
32	$d_{xxx}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3)-d_{zxy}\sin\theta_{s3}\sin(\phi_1-\phi_2)$
$\bar{4}$	$-[d_{zxx}\cos(\phi_1+\phi_2)+d_{zxy}\sin(\phi_1+\phi_2)]\sin\theta_{s3}$
$\bar{4}2m$	$-d_{zxy}\sin\theta_{s3}\sin(\phi_1+\phi_2)$
$\bar{4}3m$ (cubic)	$-d_{xyz}\sin\theta_{s3}\sin(\phi_1+\phi_2)$
23 (cubic)	$-(d_{xyz}\sin\phi_1\cos\phi_2+d_{xzy}\cos\phi_1\sin\phi_2)\sin\theta_{s3}$
432 (cubic)	$-d_{xyz}\sin\theta_{s3}\sin(\phi_1-\phi_2)$

Table 1(b). d_{eff} expressions for case (1): $o-ray(\omega_1)+o-ray(\omega_2) \rightarrow e-ray(\omega_3)$, with the assumption $d_{ijk} = d_{ikj}$.

Crystal Class	d_{eff} without the Kleinman symmetry condition	d_{eff} with the Kleinman symmetry condition
6 and 4	$d_{31} \sin\theta_{s3} \cos(\phi_1 - \phi_2)$	$d_{31} \sin\theta_{s3} \cos(\phi_1 - \phi_2)$
622 & 422	0	0
6mm & 4mm	$d_{31} \sin\theta_{s3} \cos(\phi_1 - \phi_2)$	$d_{31} \sin\theta_{s3} \cos(\phi_1 - \phi_2)$
$\bar{6}$m2	$-d_{22} \cos\theta_{s3} \sin(\phi_1 + \phi_2 + \phi_3)$	$-d_{22} \cos\theta_{s3} \sin(\phi_1 + \phi_2 + \phi_3)$
3m	$d_{31} \sin\theta_{s3} \cos(\phi_1 - \phi_2)$ $-d_{22} \cos\theta_{s3} \sin(\phi_1 + \phi_2 + \phi_3)$	$d_{31} \sin\theta_{s3} \cos(\phi_1 - \phi_2)$ $-d_{22} \cos\theta_{s3} \sin(\phi_1 + \phi_2 + \phi_3)$
$\bar{6}$	$[d_{11} \cos(\phi_1 + \phi_2 + \phi_3)$ $-d_{22} \sin(\phi_1 + \phi_2 + \phi_3)] \cos\theta_{s3}$	$[d_{11} \cos(\phi_1 + \phi_2 + \phi_3)$ $-d_{22} \sin(\phi_1 + \phi_2 + \phi_3)] \cos\theta_{s3}$
3	$[d_{11} \cos(\phi_1 + \phi_2 + \phi_3)$ $-d_{22} \sin(\phi_1 + \phi_2 + \phi_3)] \cos\theta_{s3}$ $+d_{31} \sin\theta_{s3} \cos(\phi_1 - \phi_2)$	$[d_{11} \cos(\phi_1 + \phi_2 + \phi_3)$ $-d_{22} \sin(\phi_1 + \phi_2 + \phi_3)] \cos\theta_{s3}$ $+d_{31} \sin\theta_{s3} \cos(\phi_1 - \phi_2)$
32	$d_{11} \cos\theta_{s3} \cos(\phi_1 + \phi_2 + \phi_3)$	$d_{11} \cos\theta_{s3} \cos(\phi_1 + \phi_2 + \phi_3)$
$\bar{4}$	$-[d_{31} \cos(\phi_1 + \phi_2)$ $+d_{36} \sin(\phi_1 + \phi_2)] \sin\theta_{s3}$	$-[d_{31} \cos(\phi_1 + \phi_2)$ $+d_{36} \sin(\phi_1 + \phi_2)] \sin\theta_{s3}$
$\bar{4}$2m	$-d_{36} \sin\theta_{s3} \sin(\phi_1 + \phi_2)$	$-d_{36} \sin\theta_{s3} \sin(\phi_1 + \phi_2)$
$\bar{4}$3m (cubic)	$-d_{14} \sin\theta_{s3} \sin(\phi_1 + \phi_2)$	$-d_{14} \sin\theta_{s3} \sin(\phi_1 + \phi_2)$
23 (cubic)	$-d_{14} \sin\theta_{s3} \sin(\phi_1 + \phi_2)$	$-d_{14} \sin\theta_{s3} \sin(\phi_1 + \phi_2)$
432 (cubic)	0	0

Table 2(a). d_{eff} expressions for case (2): $e-ray(\omega_1)+e-ray(\omega_2)\rightarrow o-ray(\omega_3)$, in the case of $d_{ijk}\neq d_{ikj}$, and without the Kleinman symmetry condition.

Crystal class	d_{eff} without the Kleinman symmetry condition
6 and 4	$[d_{xxz}\sin(\phi_1-\phi_3)-d_{xyz}\cos(\phi_1-\phi_3)]\cos\theta_{s1}\sin\theta_{s2}$ $+[d_{xzx}\sin(\phi_2-\phi_3)-d_{xzy}\cos(\phi_2-\phi_3)]\sin\theta_{s1}\cos\theta_{s2}$
622 & 422	$-d_{xyz}\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)-d_{xzy}\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)$
6mm & 4mm	$d_{xxz}\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+d_{xzx}\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)$
$\bar{6}$m2	$d_{yyy}\cos\theta_{s1}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)$
3m	$d_{yyy}\cos\theta_{s1}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)$ $+d_{xxz}\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+d_{xzx}\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)$
$\bar{6}$	$[d_{xxx}\sin(\phi_1+\phi_2+\phi_3)+d_{yyy}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}$
3	$[d_{xxx}\sin(\phi_1+\phi_2+\phi_3)+d_{yyy}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}$ $+[d_{xxz}\sin(\phi_1-\phi_3)-d_{xyz}\cos(\phi_1-\phi_3)]\cos\theta_{s1}\sin\theta_{s2}$ $+[d_{xzx}\sin(\phi_2-\phi_3)-d_{xzy}\cos(\phi_2-\phi_3)]\sin\theta_{s1}\cos\theta_{s2}$
32	$d_{xxx}\cos\theta_{s1}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)$ $-d_{xyz}\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)-d_{xzy}\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)$
$\bar{4}$	$[d_{xyz}\cos(\phi_1+\phi_3)-d_{xxz}\sin(\phi_1+\phi_3)]\cos\theta_{s1}\sin\theta_{s2}$ $+[d_{xzy}\cos(\phi_2+\phi_3)-d_{xzx}\sin(\phi_2+\phi_3)]\sin\theta_{s1}\cos\theta_{s2}$
$\bar{4}$2m	$d_{xyz}\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3)+d_{xzy}\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)$
$\bar{4}$3m (cubic)	$d_{xyz}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]$
23 (cubic)	$[d_{xzy}\cos\phi_1\cos\phi_3-d_{xyz}\sin\phi_1\sin\phi_3]\cos\theta_{s1}\sin\theta_{s2}$ $+[d_{xyz}\cos\phi_2\cos\phi_3-d_{xzy}\sin\phi_2\sin\phi_3]\sin\theta_{s1}\cos\theta_{s2}$
432 (cubic)	$d_{xyz}[\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)-\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)]$

Table 2(b). d_{eff} expressions for case(2) $e-\text{ray}(\omega_1)+e-\text{ray}(\omega_2)\rightarrow o-\text{ray}(\omega_3)$, with the assumption $d_{ijk}=d_{ikj}$.

Crystal Class	d_{eff} without the Kleinman symmetry condition	d_{eff} with the Kleinman symmetry condition
6 and 4	$[d_{15}\sin(\phi_1-\phi_3)-d_{14}\cos(\phi_1-\phi_3)]\cos\theta_{s1}\sin\theta_{s2}+[d_{15}\sin(\phi_2-\phi_3)-d_{14}\cos(\phi_2-\phi_3)]\sin\theta_{s1}\cos\theta_{s2}$	$d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]$
622 & 422	$-d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$	0
6mm & 4mm	$d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]$	$d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]$
$\bar{6}$m2	$d_{22}\cos\theta_{s1}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)$	$d_{22}\cos\theta_{s1}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)$
3m	$d_{22}\cos\theta_{s1}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)+d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]$	$d_{22}\cos\theta_{s1}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)+d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]$
$\bar{6}$	$[d_{11}\sin(\phi_1+\phi_2+\phi_3)+d_{22}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}$	$[d_{11}\sin(\phi_1+\phi_2+\phi_3)+d_{22}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}$
3	$[d_{11}\sin(\phi_1+\phi_2+\phi_3)+d_{22}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}+d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]-d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$	$[d_{11}\sin(\phi_1+\phi_2+\phi_3)+d_{22}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}+d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]$
32	$d_{11}\cos\theta_{s1}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)-d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$	$d_{11}\cos\theta_{s1}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)$
$\bar{4}$	$d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]-d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1+\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2+\phi_3)]$	$d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]-d_{15}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1+\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2+\phi_3)]$

$\bar{4}2m$	$d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3) + \sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]$	$d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3) + \sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]$
$\bar{4}3m$ (cubic)	$d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3) + \sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]$	$d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3) + \sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]$
23 (cubic)	$d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3) + \sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]$	$d_{14}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1+\phi_3) + \sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2+\phi_3)]$
432 (cubic)	0	0

Table 3(a). d_{eff} expressions for case (3): $o - ray(\omega_1) + e - ray(\omega_2) \rightarrow e - ray(\omega_3)$, in the case of $d_{ijk} \neq d_{ikj}$, and without the Kleinman symmetry condition

Crystal class	d_{eff} without the Kleinman symmetry condition
6 and 4	$[d_{xyz}\cos(\phi_1-\phi_3) - d_{xxz}\sin(\phi_1-\phi_3)]\sin\theta_{s2}\cos\theta_{s3}$ $- [d_{zxx}\sin(\phi_1-\phi_2) + d_{zxy}\cos(\phi_1-\phi_2)]\cos\theta_{s2}\sin\theta_{s3}$
622 & 422	$d_{xyz}\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1-\phi_3) - d_{zxy}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)$
6mm & 4mm	$- d_{xxz}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3) - d_{zxx}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$
$\bar{6}m2$	$d_{yyy}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3)$
3m	$d_{yyy}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3)$ $- d_{xxz}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3) - d_{zxx}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$
$\bar{6}$	$[d_{xxx}\sin(\phi_1+\phi_2+\phi_3) + d_{yyy}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}\cos\theta_{s3}$
3	$[d_{xxx}\sin(\phi_1+\phi_2+\phi_3) + d_{yyy}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}\cos\theta_{s3}$ $- [d_{xxz}\sin(\phi_1-\phi_3) - d_{xyz}\cos(\phi_1-\phi_3)]\sin\theta_{s2}\cos\theta_{s3}$ $- [d_{zxx}\sin(\phi_1-\phi_2) + d_{zxy}\cos(\phi_1-\phi_2)]\cos\theta_{s2}\sin\theta_{s3}$
32	$d_{xxx}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$ $+ d_{xyz}\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1-\phi_3) - d_{zxy}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)$
$\bar{4}$	$[d_{xyz}\cos(\phi_1+\phi_3) - d_{xxz}\sin(\phi_1+\phi_3)]\sin\theta_{s2}\cos\theta_{s3}$ $+ [d_{zxy}\cos(\phi_1+\phi_2) - d_{zxx}\sin(\phi_1+\phi_2)]\cos\theta_{s2}\sin\theta_{s3}$
$\bar{4}2m$	$d_{xyz}\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_3) + d_{zxy}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1+\phi_2)$
$\bar{4}3m$ (cubic)	$d_{xyz}[\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1+\phi_2)]$
23 (cubic)	$[d_{xyz}\cos\phi_1\cos\phi_3 - d_{xzy}\sin\phi_1\sin\phi_3]\sin\theta_{s2}\cos\theta_{s3}$ $+ [d_{xzy}\cos\phi_1\cos\phi_2 - d_{xyz}\sin\phi_1\sin\phi_2]\cos\theta_{s2}\sin\theta_{s3}$
432 (cubic)	$d_{xyz}[\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1-\phi_3) - \cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)]$

Table 3(b). d_{eff} expressions for case (3) $o-\text{ray}(\omega_1)+e-\text{ray}(\omega_2)\rightarrow e-\text{ray}(\omega_3)$, with the assumption $d_{ijk}=d_{ikj}$.

Crystal Class	d_{eff} without the Kleinman symmetry condition	d_{eff} with the Kleinman symmetry condition
6 and 4	$[d_{14}\cos(\phi_1-\phi_3) - d_{15}\sin(\phi_1-\phi_3)]\sin\theta_{s2}\cos\theta_{s3} - d_{31}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$	$-d_{31}[\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)]$
622 & 422	$d_{14}\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1-\phi_3)$	0
6mm & 4mm	$-d_{15}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3) - d_{31}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$	$-d_{31}[\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)]$
$\bar{6}$m2	$d_{22}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3)$	$d_{22}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3)$
3m	$d_{22}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3) - d_{15}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3) - d_{31}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$	$d_{22}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3) - d_{31}[\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)]$
$\bar{6}$	$[d_{11}\sin(\phi_1+\phi_2+\phi_3) + d_{22}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}\cos\theta_{s3}$	$[d_{11}\sin(\phi_1+\phi_2+\phi_3) + d_{22}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}\cos\theta_{s3}$
3	$[d_{11}\sin(\phi_1+\phi_2+\phi_3) + d_{22}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}\cos\theta_{s3} + [d_{14}\cos(\phi_1-\phi_3) - d_{15}\sin(\phi_1-\phi_3)]\sin\theta_{s2}\cos\theta_{s3} - d_{31}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$	$[d_{11}\sin(\phi_1+\phi_2+\phi_3) + d_{22}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}\cos\theta_{s3} - d_{31}[\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)]$
32	$d_{11}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3) + d_{14}\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1-\phi_3)$	$d_{11}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$
$\bar{4}$	$[d_{14}\cos(\phi_1+\phi_3) - d_{15}\sin(\phi_1+\phi_3)]\sin\theta_{s2}\cos\theta_{s3} + [d_{36}\cos(\phi_1+\phi_2) - d_{31}\sin(\phi_1+\phi_2)]\cos\theta_{s2}\sin\theta_{s3}$	$d_{14}[\cos(\phi_1+\phi_3)\sin\theta_{s2}\cos\theta_{s3} + \cos(\phi_1+\phi_2)\cos\theta_{s2}\sin\theta_{s3}] - d_{31}[\sin(\phi_1+\phi_3)\sin\theta_{s2}\cos\theta_{s3} + \sin(\phi_1+\phi_2)\cos\theta_{s2}\sin\theta_{s3}]$
$\bar{4}$2m	$d_{14}\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_3) + d_{36}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1+\phi_2)$	$d_{14}[\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1+\phi_2)]$

$\bar{4}3m$ (cubic)	$d_{14}[\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1+\phi_2)]$	$d_{14}[\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1+\phi_2)]$
23 (cubic)	$d_{14}[\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1+\phi_2)]$	$d_{14}[\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_3) + \cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1+\phi_2)]$
432 (cubic)	0	0

Table 4(a). d_{eff} expressions for case (4): $o-ray(\omega_1)+e-ray(\omega_2) \rightarrow o-ray(\omega_3)$, in the case of $d_{ijk} \neq d_{ikj}$, and without the Kleinman symmetry condition.

Crystal class	d_{eff} without the Kleinman symmetry condition
6 and 4	$(d_{xxz}\cos(\phi_1-\phi_3)+d_{xyz}\sin(\phi_1-\phi_3))\sin\theta_{s2}$
622 & 422	$d_{xyz}\sin\theta_{s2}\sin(\phi_1-\phi_3)$
6mm & 4mm	$d_{xxz}\sin\theta_{s2}\cos(\phi_1-\phi_3)$
$\bar{6}m2$	$-d_{yyy}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)$
3m	$d_{xxz}\sin\theta_{s2}\cos(\phi_1-\phi_3)-d_{yyy}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)$
$\bar{6}$	$(d_{xxx}\cos(\phi_1+\phi_2+\phi_3)-d_{yyy}\sin(\phi_1+\phi_2+\phi_3))\cos\theta_{s2}$
3	$[d_{xxx}\cos(\phi_1+\phi_2+\phi_3)-d_{yyy}\sin(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}$ $+[d_{xxz}\cos(\phi_1-\phi_3)+d_{xyz}\sin(\phi_1-\phi_3)]\sin\theta_{s2}$
32	$d_{xxx}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)+d_{xyz}\sin\theta_{s2}\sin(\phi_1-\phi_3)$
$\bar{4}$	$-[d_{xxz}\cos(\phi_1+\phi_3)+d_{xyz}\sin(\phi_1+\phi_3)]\sin\theta_{s2}$
$\bar{4}2m$	$-d_{xyz}\sin\theta_{s2}\sin(\phi_1+\phi_3)$
$\bar{4}3m$ (cubic)	$-d_{xyz}\sin\theta_{s2}\sin(\phi_1+\phi_3)$
23 (cubic)	$-(d_{xzy}\sin\phi_1\cos\phi_3+d_{xyz}\cos\phi_1\sin\phi_3)\sin\theta_{s2}$
432 (cubic)	$d_{xyz}\sin\theta_{s2}\sin(\phi_1-\phi_3)$

Table 4(b). d_{eff} expressions for case (4): $o-\mathrm{ray}(\omega_1)+e-\mathrm{ray}(\omega_2)\rightarrow o-\mathrm{ray}(\omega_3)$, with the assumption $d_{ijk}=d_{ikj}$.

Crystal Class	d_{eff} without the Kleinman symmetry condition	d_{eff} with the Kleinman symmetry condition
6 and 4	$[d_{15}\cos(\phi_1-\phi_3)$ $+ d_{14}\sin(\phi_1-\phi_3)]\sin\theta_{s2}$	$d_{15}\sin\theta_{s2}\cos(\phi_1-\phi_3)$
622 & 422	$d_{14}\sin\theta_{s2}\sin(\phi_1-\phi_3)$	0
6mm & 4mm	$d_{15}\sin\theta_{s2}\cos(\phi_1-\phi_3)$	$d_{15}\sin\theta_{s2}\cos(\phi_1-\phi_3)$
$\bar{6}$m2	$-d_{22}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)$	$-d_{22}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)$
3m	$d_{15}\sin\theta_{s2}\cos(\phi_1-\phi_3)$ $-d_{22}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)$	$d_{15}\sin\theta_{s2}\cos(\phi_1-\phi_3)$ $-d_{22}\cos\theta_{s2}\sin(\phi_1+\phi_2+\phi_3)$
$\bar{6}$	$[d_{11}\cos(\phi_1+\phi_2+\phi_3)$ $-d_{22}\sin(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}$	$[d_{11}\cos(\phi_1+\phi_2+\phi_3)$ $-d_{22}\sin(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}$
3	$[d_{11}\cos(\phi_1+\phi_2+\phi_3)$ $-d_{22}\sin(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}$ $+[d_{15}\cos(\phi_1-\phi_3)$ $+d_{14}\sin(\phi_1-\phi_3)]\sin\theta_{s2}$	$[d_{11}\cos(\phi_1+\phi_2+\phi_3)$ $-d_{22}\sin(\phi_1+\phi_2+\phi_3)]\cos\theta_{s2}$ $+d_{15}\sin\theta_{s2}\cos(\phi_1-\phi_3)$
32	$d_{11}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)$ $+d_{14}\sin\theta_{s2}\sin(\phi_1-\phi_3)$	$d_{11}\cos\theta_{s2}\cos(\phi_1+\phi_2+\phi_3)$
$\bar{4}$	$-[d_{15}\cos(\phi_1+\phi_3)$ $+d_{14}\sin(\phi_1+\phi_3)]\sin\theta_{s2}$	$-[d_{15}\cos(\phi_1+\phi_3)$ $+d_{14}\sin(\phi_1+\phi_3)]\sin\theta_{s2}$
$\bar{4}$2m	$-d_{14}\sin\theta_{s2}\sin(\phi_1+\phi_3)$	$-d_{14}\sin\theta_{s2}\sin(\phi_1+\phi_3)$
$\bar{4}$3m (cubic)	$-d_{14}\sin\theta_{s2}\sin(\phi_1+\phi_3)$	$-d_{14}\sin\theta_{s2}\sin(\phi_1+\phi_3)$
23 (cubic)	$-d_{14}\sin\theta_{s2}\sin(\phi_1+\phi_3)$	$-d_{14}\sin\theta_{s2}\sin(\phi_1+\phi_3)$
$\bar{4}$2m (cubic)	0	0

Table 5(a). deff expressions for case (5): $e-\text{ray}(\omega_1)+e-\text{ray}(\omega_2)\rightarrow e-\text{ray}(\omega_3)$, in the case of $d_{ijk}\neq d_{ikj}$, and without the Kleinman symmetry condition.

Crystal class	deff without the Kleinman symmetry condition
6 and 4	$[d_{xxz}\cos(\phi_1-\phi_3)+d_{xyz}\sin(\phi_1-\phi_3)]\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}$ $+[d_{xzx}\cos(\phi_2-\phi_3)+d_{xzy}\sin(\phi_2-\phi_3)]\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+[d_{zxx}\cos(\phi_1-\phi_2)-d_{zxy}\sin(\phi_1-\phi_2)]\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}$ $+d_{zzz}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
622 & 422	$d_{xyz}\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3)$ $+d_{xzy}\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2-\phi_3)$ $-d_{zxy}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$
6mm & 4mm	$d_{xxz}\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1-\phi_3)$ $+d_{xzx}\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_2-\phi_3)$ $+d_{zxx}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)+d_{zzz}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
$\bar{6}$m2	$d_{yyy}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$
3m	$d_{yyy}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$ $+d_{xxz}\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\cos(\phi_1-\phi_3)$ $+d_{xzx}\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_2-\phi_3)$ $+d_{zxx}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)+d_{zzz}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
$\bar{6}$	$[d_{yyy}\sin(\phi_1+\phi_2+\phi_3)-d_{xxx}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$
3	$[d_{yyy}\sin(\phi_1+\phi_2+\phi_3)-d_{xxx}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+[d_{xxz}\cos(\phi_1-\phi_3)+d_{xyz}\sin(\phi_1-\phi_3)]\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}$ $+[d_{xzx}\cos(\phi_2-\phi_3)+d_{xzy}\sin(\phi_2-\phi_3)]\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+[d_{zxx}\cos(\phi_1-\phi_2)-d_{zxy}\sin(\phi_1-\phi_2)]\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}$ $+d_{zzz}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
32	$-d_{xxx}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3)$ $+d_{xyz}\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3)$ $+d_{xzy}\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2-\phi_3)$ $-d_{zxy}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$
$\bar{4}$	$[d_{xxz}\cos(\phi_1+\phi_3)+d_{xyz}\sin(\phi_1+\phi_3)]\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}$ $+[d_{xzx}\cos(\phi_2+\phi_3)+d_{xzy}\sin(\phi_2+\phi_3)]\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+[d_{zxx}\cos(\phi_1+\phi_2)+d_{zxy}\sin(\phi_1+\phi_2)]\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}$

$\bar{4}2m$	$d_{xyz}\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_3)$ $+ d_{xzy}\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2+\phi_3)$ $+ d_{zxy}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1+\phi_2)$
$\bar{4}3m$ (cubic)	$d_{xyz}[\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_3)$ $+ \sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2+\phi_3) + \cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1+\phi_2)]$
23 (cubic)	$[d_{xyz}\sin\phi_1\cos\phi_3 + d_{xzy}\cos\phi_1\sin\phi_3)]\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}$ $+ [d_{xyz}\cos\phi_2\sin\phi_3 + d_{xzy}\sin\phi_2\cos\phi_3)]\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+ [d_{xyz}\cos\phi_1\sin\phi_2 + d_{xzy}\sin\phi_1\cos\phi_2)]\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}$
432 (cubic)	$d_{xyz}\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1-\phi_3)$ $- d_{xyz}\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2-\phi_3)$ $- d_{xyz}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1-\phi_2)$

Table 5(b). d_{eff} expressions for case (5): $e-ray(\omega_1)+e-ray(\omega_2)\rightarrow e-ray(\omega_3)$, in the case of $d_{ijk}=d_{ikj}$, but without the Kleinman symmetry condition.

Crystal class	d_{eff} without the Kleinman symmetry condition
6 and 4	$[d_{15}\cos(\phi_1-\phi_3)+d_{14}\sin(\phi_1-\phi_3)]\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}$ $+[d_{15}\cos(\phi_2-\phi_3)+d_{14}\sin(\phi_2-\phi_3)]\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+d_{31}\cos(\phi_1-\phi_2)\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}$ $+d_{33}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
622 & 422	$d_{14}\cos\theta_{s3}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]$
6mm & 4mm	$d_{15}\cos\theta_{s3}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$ $+d_{31}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)+d_{33}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
$\bar{6}m2$	$d_{22}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$
3m	$d_{22}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$ $+d_{15}\cos\theta_{s3}[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$ $+d_{31}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)+d_{33}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
$\bar{6}$	$[d_{22}\sin(\phi_1+\phi_2+\phi_3)-d_{11}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$
3	$[d_{22}\sin(\phi_1+\phi_2+\phi_3)-d_{11}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+[d_{15}\cos(\phi_1-\phi_3)+d_{14}\sin(\phi_1-\phi_3)]\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}$ $+[d_{15}\cos(\phi_2-\phi_3)+d_{14}\sin(\phi_2-\phi_3)]\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+d_{31}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)$ $+d_{33}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
32	$-d_{11}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3)$ $+d_{14}\cos\theta_{s3}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2-\phi_3)]$
$\bar{4}$	$[d_{15}\cos(\phi_1+\phi_3)+d_{14}\sin(\phi_1+\phi_3)]\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}$ $+[d_{15}\cos(\phi_2+\phi_3)+d_{14}\sin(\phi_2+\phi_3)]\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+[d_{31}\cos(\phi_1+\phi_2)+d_{36}\sin(\phi_1+\phi_2)]\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}$
$\bar{4}2m$	$d_{14}\cos\theta_{s3}[\cos\theta_{s1}\sin\theta_{s2}\sin(\phi_1+\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\sin(\phi_2+\phi_3)]$ $+d_{36}\cos\theta_{s1}\cos\theta_{s1}\sin\theta_{s3}\sin(\phi_1+\phi_2)$
$\bar{4}3m$ (cubic)	$d_{14}[\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2+\phi_3)+\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_3)$ $+\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1+\phi_2)]$
23 (cubic)	$d_{14}[\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2+\phi_3)+\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_3)$ $+\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1+\phi_2)]$
432 (cubic)	0

Table 5(c). d_{eff} expressions for case (5): $e-\mathrm{ray}(\omega_1)+e-\mathrm{ray}(\omega_2)\rightarrow e-\mathrm{ray}(\omega_3)$, with the Kleinman symmetry condition.

Crystal class	d_{eff} with the Kleinman symmetry condition
6 and 4	$d_{15}\cos\theta_{s3}\,[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$ $+\,d_{31}\cos(\phi_1-\phi_2)\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}$ $+\,d_{33}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
622 & 422	0
6mm & 4mm	$d_{31}\cos\theta_{s3}\,[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$ $+\,d_{31}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)+d_{33}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
$\bar{6}$m2	$d_{22}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$
3m	$d_{22}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_2+\phi_3)$ $+\,d_{31}\cos\theta_{s3}\,[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$ $+\,d_{31}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)+d_{33}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
$\bar{6}$	$[\,d_{22}\sin(\phi_1+\phi_2+\phi_3)-d_{11}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$
3	$[\,d_{22}\sin(\phi_1+\phi_2+\phi_3)-d_{11}\cos(\phi_1+\phi_2+\phi_3)]\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+\,d_{31}\cos\theta_{s3}\,[\cos\theta_{s1}\sin\theta_{s2}\cos(\phi_1-\phi_3)+\sin\theta_{s1}\cos\theta_{s2}\cos(\phi_2-\phi_3)]$ $+\,d_{31}\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\cos(\phi_1-\phi_2)$ $+\,d_{33}\sin\theta_{s1}\sin\theta_{s2}\sin\theta_{s3}$
32	$-\,d_{11}\cos\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\cos(\phi_1+\phi_2+\phi_3)$
$\bar{4}$	$[d_{31}\cos(\phi_1+\phi_3)+d_{14}\sin(\phi_1+\phi_3)]\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}$ $+\,[d_{31}\cos(\phi_2+\phi_3)+d_{14}\sin(\phi_2+\phi_3)]\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}$ $+\,[d_{31}\cos(\phi_1+\phi_2)+d_{14}\sin(\phi_1+\phi_2)]\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}$
$\bar{4}$2m	$d_{14}\,[\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2+\phi_3)+\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_3)$ $+\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1+\phi_2)]$
$\bar{4}$3m (cubic)	$d_{14}\,[\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2+\phi_3)+\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_3)$ $+\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1+\phi_2)]$
23 (cubic)	$d_{14}\,[\sin\theta_{s1}\cos\theta_{s2}\cos\theta_{s3}\sin(\phi_2+\phi_3)+\cos\theta_{s1}\sin\theta_{s2}\cos\theta_{s3}\sin(\phi_1+\phi_3)$ $+\cos\theta_{s1}\cos\theta_{s2}\sin\theta_{s3}\sin(\phi_1+\phi_2)]$
432 (cubic)	0

Table 6. d_{eff} expressions for case (6): $o-ray(\omega_1)+o-ray(\omega_2)\rightarrow o-ray(\omega_3)$.

Crystal Class	d_{eff} for the case of $d_{ijk} \neq d_{ikj}$ and without the Kleinman symmetry condition	d_{eff} for the case of $d_{ijk} = d_{ikj}$ or with the Kleinman symmetry condition
6 and 4	0	0
622 & 422	0	0
6mm & 4mm	0	0
$\bar{6}m2$	$-d_{yyy}\cos(\phi_1+\phi_2+\phi_3)$	$-d_{22}\cos(\phi_1+\phi_2+\phi_3)$
3m	$-d_{yyy}\cos(\phi_1+\phi_2+\phi_3)$	$-d_{22}\cos(\phi_1+\phi_2+\phi_3)$
$\bar{6}$	$-d_{xxx}\sin(\phi_1+\phi_2+\phi_3)$ $-d_{yyy}\cos(\phi_1+\phi_2+\phi_3)$	$-d_{11}\sin(\phi_1+\phi_2+\phi_3)$ $-d_{22}\cos(\phi_1+\phi_2+\phi_3)$
3	$-d_{xxx}\sin(\phi_1+\phi_2+\phi_3)$ $-d_{yyy}\cos(\phi_1+\phi_2+\phi_3)$	$-d_{11}\sin(\phi_1+\phi_2+\phi_3)$ $-d_{22}\cos(\phi_1+\phi_2+\phi_3)$
32	$-d_{xxx}\sin(\phi_1+\phi_2+\phi_3)$	$-d_{11}\sin(\phi_1+\phi_2+\phi_3)$
$\bar{4}$	0	0
$\bar{4}2m$	0	0
$\bar{4}3m$ (cubic)	0	0
23 (cubic)	0	0
432 (cubic)	0	0